

Statistics of an adiabatic charge pump

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We investigate the effect of time-dependent cyclic-adiabatic driving on the charge transport in quantum junction. We propose a nonequilibrium Greens function formalism to study statistics of the charge pumped (at zero bias) through the junction. The formulation is used to demonstrate charge pumping in a single electronic level coupled to two (electronic) reservoirs with time dependent couplings. Analytical expression for the average pumped current for a general cyclic driving is derived. It is found that for zero bias, for a certain class of driving, the Berry phase contributes only to the odd cumulants. To contrast, a quantum master equation formulation does not show Berry-phase effect at all.

It is well known that the effect of adiabatically varying few parameters in the Hamiltonian in a cyclic manner enters in the wavefunction in the form of a phase factor. This phase factor consists of two parts, one is called dynamical part (which, in general, depends on how fast the parameters are varied) and the second one, generally known as Berry phase (also called geometric phase) that depends only on the path (area) traced in the parameter space and independent of how fast it is traced provided adiabatic condition is satisfied[1–3]. Somewhat counter-intuitive, this phase factor may lead to changes in macroscopic observables, like finite spin or charge currents in one dimensional phase coherent rings at equilibrium[4–6]. Originally developed in the context of closed quantum systems, recent works have extended the geometric phase concept to the case of open quantum systems out of equilibrium[7, 8]. This is usually treated within the quantum master equation (QME) approach[9]. Stochastic variation of system parameters is known to induce net flux in open systems like quantum heat pumps[10, 11], quantum electron pumps[12] and also classical stochastic systems like enzyme kinetics, molecular motors and living cell locomotion[13]. On a similar footing, adiabatic cyclic variation of parameters in the Hamiltonian may also lead to finite flux [14]. Switkes et.al.,[15] have experimentally demonstrated an adiabatic quantum pump by modulating confining potential of an open quantum dot in a cyclic manner, leading to a finite voltage drop across the quantum dot. Modifying the potential at two ends of the dot changes the character of the wavefunction and therefore modifies the couplings to the electron reservoirs. In this work we explore this aspect within the most general framework based on non-equilibrium Greens function [16]. The adiabatic driving may also effect the statistics of charge transfer and the steady-state fluctuation relation due to Gallavoti Cohen (GC) type symmetry [17] may also get modified. Ren et.al. [10] have recently used QME to study heat pumping and fluctuations of heat transfer in a two-level system sandwiched between two thermal reservoirs. It was shown that in the case of time-dependent temperature modulations of the two heat reservoirs, heat transfer statistics

does not admit GC type symmetry. It was also argued that modulating couplings to thermal reservoirs does not lead to any pumping. Several methods like scattering theory[18, 19], Floquet scattering theory[20], adiabatic master equation approach[8, 12] etc., have been developed for studying the statistics of adiabatic pumping. But each of these methods has its own advantages and short-comings. In this work we develop a scheme within NEGF formalism (which in principle is exact) and apply it to study the effect of cyclic-adiabatic driving on the charge transfer statistics in a resonant level model. We find that finite charge transfer between two reservoirs (at the same thermodynamic states) connected through a single level quantum system is possible by modulating the couplings to the reservoirs in an adiabatic-cyclic manner. We present an analytic expression for the pumped current for a general driving. The direction of the net charge flow can be varied by changing the sign of the phase difference between the two time-dependent couplings. We find that the flux direction also depends on the energy difference between the chemical potentials (μ) of reservoirs and the level energy ($\mu - \omega_0$). In general, the charge transfer fluctuations are modified due to the Berry phase. However, at equilibrium, for certain class of drivings, asymmetric fluctuations (odd cumulants) are generated solely due to the Berry-phase. The full statistics of the pumped charge satisfy a steady-state fluctuation relation. We emphasize that the present formulation shows that it is possible to pump a finite net charge in *noninteracting* open quantum junctions, unlike a simple QME formulation which does not lead to any pumping due to the Berry-phase [10, 12].
Model Hamiltonian: A general Hamiltonian for the description of electron transport in a quantum junction where a molecular system is coupled to two (non interacting) electronic reservoirs is

$$\hat{H}(t) = \sum_r \epsilon_r(t) d_r^\dagger d_r + H_{int} + \sum_{\alpha k} \epsilon_{\alpha,k} c_{\alpha k}^\dagger c_{\alpha k} + \sum_{r, \alpha k} [g_{\alpha k, r}^*(t) c_{\alpha k}^\dagger d_r + g_{\alpha k, r}(t) d_r^\dagger c_{\alpha k}] \quad (1)$$

where $d_r^\dagger(d_r)$ stands for electron creation (annihilation)

operator in the r -th system orbital while $c_{\alpha,k}^\dagger$ ($c_{\alpha,k}$) is for electron creation (annihilation) operator on the left or right ($\alpha = L/R$) reservoir in the energy state $\epsilon_{\alpha,k}$. H_{int} is the Hamiltonian to account for all other possible interactions in the system, like Coulomb and electron-phonon interactions. Here system lead couplings and/or single electron orbital energies can be periodically modulated (which can be experimentally realized by applying time dependent gate voltages). In this work we consider the case when the driving time period is large compared to the internal relaxation time scales in the system such that the system at any time is at steady state with respect to reservoirs.

Two-point measurement: Under these conditions, we consider two simultaneous measurements of electron number in the left and the right reservoirs (as $[N_L, N_R] = 0$, simultaneous measurement is quantum mechanically allowed) at time T_0 and $T > T_0$. The probability of change in the number of particles, $N_\alpha(T) - N_\alpha(T_0)$, in the left and the right reservoirs during the measurement time, $T - T_0$, can be computed using the generating function (GF) containing corresponding counting parameters λ_L and λ_R as

$$P(n_L, n_R, T - T_0) = \int_0^{2\pi} \frac{d\lambda_L}{2\pi} \int_0^{2\pi} \frac{d\lambda_R}{2\pi} \mathcal{Z}(\lambda_L, \lambda_R, T - T_0) e^{i(\lambda_L n_L + \lambda_R n_R)} \quad (2)$$

Using the two time quantum measurement formalism the generating function for particle number counting on both reservoirs can be written as [17]

$$\mathcal{Z}(\lambda, T - T_0) = \langle e^{-i(\lambda_L N_L(T) + \lambda_R N_R(T))} e^{i(\lambda_L N_L(T_0) + \lambda_R N_R(T_0))} \rangle_{\rho(T_0)} \quad (3)$$

where $\rho(T_0)$ is the density matrix of the system + reservoirs at time T_0 and $N_L = \sum_k c_{Lk}^\dagger c_{Lk}$, $N_R = \sum_k c_{Rk}^\dagger c_{Rk}$ are the particle number operators corresponding to left and right reservoirs respectively. Assuming $[\rho(T_0), N_L] = 0$ and $[\rho(T_0), N_R] = 0$ (or, more generally, system and reservoirs are decoupled at time T_0 and each are at equilibrium independently), Eq.(3) can be recast as

$$\begin{aligned} \mathcal{Z}(\lambda_L, \lambda_R, T - T_0) &= \left\langle \mathcal{U}_{(\frac{\lambda_L}{2}, \frac{\lambda_R}{2})}(T_0, T) \mathcal{U}_{(-\frac{\lambda_L}{2}, -\frac{\lambda_R}{2})}(T, T_0) \right\rangle_{\rho(T_0)} \\ &= \left\langle \mathcal{T}_c e^{-\frac{i}{\hbar} \int_c H_T^{\lambda(\tau)}(\tau) d\tau} \right\rangle_{\rho(T_0)} \end{aligned} \quad (4)$$

where in the last line time dependent $\lambda(\tau)$ has been defined on the Keldysh contour[21, 22] (which goes from T_0 to T and back to T_0)[23] as $\lambda(\tau) = (-\frac{\lambda_L}{2}, -\frac{\lambda_R}{2})$ on the forward contour and $\lambda(\tau) = (\frac{\lambda_L}{2}, \frac{\lambda_R}{2})$ on the backward contour. Where \mathcal{T}_c refers to time ordering operator on the Keldysh contour. The evolution on the Keldysh contour is with respect to $\lambda(\tau)$ dependent Hamiltonian (note

that λ -dependent evolution is no longer unitary), and evolves the ket and the bra with different λ -dependent Hamiltonian [24], which can be obtained by replacing the last line in Eq. (1) by $\sum_{r,\alpha k} [g_{\alpha k,r}^*(\tau) e^{i\lambda_\alpha(\tau)} c_{\alpha k}^\dagger d_r + g_{\alpha k,r}(\tau) e^{-i\lambda_\alpha(\tau)} d_r^\dagger c_{\alpha k}]$. Hence the effect of measurement is reflected in the form of modified (λ -dependent) couplings to the reservoirs[25].

GF in terms of NEGF: Taking λ_R derivative [23] of the logarithm of Eq.(4), we get

$$\begin{aligned} \left[\frac{\partial \ln \mathcal{Z}(\lambda_L, \lambda_R, T - T_0)}{\partial(i\lambda_R)} \right] &= \frac{1}{2} \sum_{k,r} \int_{T_0}^T dt_1 \\ &[g_{Rk,r}^*(t_1) e^{-i\frac{\lambda_R}{2}} G_{r,Rk}^{++}(t_1, t_1) - g_{Rk,r}(t_1) e^{i\frac{\lambda_R}{2}} G_{Rk,r}^{++}(t_1, t_1) \\ &+ g_{Rk,r}^*(t_1) e^{i\frac{\lambda_R}{2}} G_{r,Rk}^{--}(t_1, t_1) - g_{Rk,r}(t_1) e^{-i\frac{\lambda_R}{2}} G_{Rk,r}^{--}(t_1, t_1)] \end{aligned} \quad (5)$$

where $G_{Rk,r}^{++}(t, t')$, $G_{r,Rk}^{++}(t, t')$, $G_{Rk,r}^{--}(t, t')$ and $G_{r,Rk}^{--}(t, t')$ are appropriate real time projections of mixed contour ordered Greens functions between system orbitals and reservoir states defined as $G_{Rk,r}^c(\tau, \tau') = -\frac{i}{\hbar} \langle \mathcal{T}_c c_{Rk}(\tau) d_r^\dagger(\tau') \rangle$ and $G_{r,Rk}^c(\tau, \tau') = -\frac{i}{\hbar} \langle \mathcal{T}_c d_r(\tau) c_{Rk}^\dagger(\tau') \rangle$. Here '+' ('-') index refers to the time variable located on the upper (lower) Keldysh contour[21]. Equation (5) can be recast in terms of the system Greens function matrix alone [22] which can be expressed in terms of Wigner transformed quantities [16]. In the large measurement time limit, Eq. (5) can be recast as [26],

$$\begin{aligned} \left[\frac{\partial \ln \mathcal{Z}(\lambda_L, \lambda_R, T - T_0)}{\partial(i\lambda_R)} \right] &= \int_{T_0}^T dt \int_{-\infty}^{+\infty} \frac{d\omega}{2\pi} \\ \text{Tr} \left[\check{\Sigma}_R^{+-}(\omega, t) \check{\mathbb{G}}^{-+}(\omega, t) - \check{\mathbb{G}}^{+-}(\omega, t) \check{\Sigma}_R^{+-}(\omega, t) \right] \end{aligned} \quad (6)$$

Here the trace is over all the system orbitals and $\Sigma_R^{+-}(\omega, t)$, etc. are (Wigner transformed) self-energy matrices due to interaction with the right reservoir. $G^{+-}(\omega, t)$, etc. are Wigner transforms of real time projections of system contour ordered Greens function matrices with elements

$$G_{mn}^c(\tau, \tau') = -\frac{i}{\hbar} \langle \mathcal{T}_c d_m(\tau) d_n^\dagger(\tau') \rangle \quad (7)$$

where $\langle \dots \rangle$ is the average with respect to the density matrix evolving on Keldysh contour as defined in Eq.(4). In Eq. (6) we have used the notation $\check{A}^{-\eta} = -A^{-\eta}$ and $\check{A}^{+\eta} = A^{+\eta}$, where $\eta = \pm$, to simplify the following expressions. Expression for $\left[\frac{\partial \ln \mathcal{Z}(\lambda_L, \lambda_R, T - T_0)}{\partial(i\lambda_L)} \right]$ can be obtained by replacing 'R' with 'L' in Eq.(5). Hence all that we have to do to get the final expression for the GF is to calculate the $\lambda(\tau)$ -dependent Greens functions appearing in Eq. (6). This is done in the following.

Approximation for the Greens function: The λ -dependent Greens function matrix defined on Keldysh

contour (with matrix elements in system orbital space defined in Eq.(7)) satisfy the following equation of motion [22] (also known as left-Dyson equation) on the Keldysh contour

$$\int_c d\tau_1 [G_0^{-1}(\tau, \tau_1) - \Sigma^c(\tau, \tau_1)] G^c(\tau_1, \tau') = \delta^c(\tau, \tau') \quad (8)$$

where $\left[i\hbar \frac{\partial}{\partial \tau} - H_0(\tau) \right] G_0(\tau, \tau') = \delta^c(\tau, \tau')$ and $\Sigma^c(\tau, \tau') = \Sigma_{int}^c(\tau, \tau') + \Sigma_{leads}^c(\tau, \tau')$ is the total self energy due to H_{int} and system-reservoir coupling. $\Sigma_{leads}^c(\tau, \tau')$ has matrix elements,

$$\begin{aligned} \Sigma_{leads, rr'}^c(\tau, \tau') &= \sum_{\alpha} \sum_{k, k'} g_{\alpha k, r}(\tau) e^{-i(\lambda_{\alpha}(\tau) - \lambda_{\alpha}(\tau'))} \\ &\times G_{\alpha k, \alpha k'}^0(\tau, \tau') g_{\alpha k', r'}^*(\tau'). \end{aligned} \quad (9)$$

Here $G_{\alpha k, \alpha k'}^0(\tau, \tau')$ is contour ordered Greens functions of free reservoir. An explicit expression for Σ_{int}^c depends on H_{int} which we keep general. The above equation can be projected onto the real times to obtain equations of motion for Keldysh matrix $\check{G}(t, t')$. By performing Wigner transformation followed by Fourier transformation over quantum time, Eq.(10) can be recast as [21]

$$\begin{aligned} \check{G}(\omega, t) &= \check{G}_{ad}(\omega, t) + \check{G}_{ad}(\omega, t) \left[\check{G}_{ad}^{-1}(\omega, t) \right. \\ &\left. \left(\mathbb{I} - \exp \left[-\frac{i}{2} [\overleftarrow{\partial}_t \overrightarrow{\partial}_\omega - \overleftarrow{\partial}_\omega \overrightarrow{\partial}_t] \right] \check{G}(\omega, t) \right) \right] \end{aligned} \quad (10)$$

where \mathbb{I} is the identity matrix and $\check{G}(\omega, t)$ is the system Greens function matrix Fourier transformed over quantum time. $\overleftarrow{\partial}_t, \overrightarrow{\partial}_t$ represent classical time t derivative acting on the function to its left or to its right respectively, similarly $\overleftarrow{\partial}_\omega, \overrightarrow{\partial}_\omega$ represent ' ω ' derivatives. The adiabatic contribution, \check{G}_{ad} , to the Greens function satisfies the matrix equation,

$$\check{G}_{ad}(\omega, t) = \left[\check{G}_0^{-1}(\omega, t) - \check{\Sigma}(\omega, t) \right]^{-1}. \quad (11)$$

This is similar to the usual steady state Greens function but the parameters are replaced with time dependent parameters. Here $\check{\Sigma}(\omega, t)$ and $\check{G}_0(\omega, t)$ are, respectively, the self energy and the non interacting system Greens function matrices. The second term in Eq.(10) represents a correction due to time variation of the parameters. Note that Eq. (10) is exact. We solve Eq.(10) to lowest order in time derivative by iterating the equation for \check{G} perturbatively in terms of time derivatives of \check{G}_{ad} and retaining only terms linear in first derivative in (classical) time.

$$\check{G}(\omega, t) = \check{G}_{ad}(\omega, t) + \frac{i}{2} \check{G}_{ad}(\omega, t) \{ \check{G}_{ad}^{-1}(\omega, t), \check{G}_{ad}(\omega, t) \}. \quad (12)$$

where $\{, \}$ stand for Poisson bracket in t and ω variables. We note that both the left- and the right-Dyson equations lead to the same approximate Eq. (12). We also

note that Eq. (12) for $\lambda = 0$ case preserves all the symmetries (as a consequence of the unitary evolution) of the Greens functions [26]. Equation (11) and Eq. (12) can be solved to obtain λ -dependent Greens functions which can be used to compute charge transfer statistics using Eq.(6). In the following, we apply this formalism to study Berry effect on the charge transfer statistics in a resonant level model.

Adiabatically driven resonant level model: Consider a single electronic site connected via time-dependent hopping to two electronic reservoirs. The Hamiltonian describing this model is the same as in Eq. (1) with $H_{int}(t) = 0$ and only single system orbital with time independent energy $\epsilon = \omega_0$. We put $\hbar = 1$ in this section. We compute the adiabatic Greens functions and the lowest order non-adiabatic corrections using Eq.(11) and Eq. (12) [26]. Using this Greens functions in Eq.(9),

we obtain an expression for $\frac{1}{(T-T_0)} \left[\frac{\partial \ln \mathcal{Z}(\lambda_L, \lambda_R, T-T_0)}{\partial (i\lambda_R)} \right]$ [26]. In order to emphasize the Berry phase effect, we consider the two reservoirs at the same thermodynamic equilibrium ($\mu_L = \mu_R = \mu$ i.e., zero external bias and inverse temperatures, $\beta_L = \beta_R = \beta$). From n_L and n_R measurements, we can obtain the statistics of net charge, $n = (n_L - n_R)/2$, transferred between the two reservoirs and the change in the charge, $N = n_L + n_R$, on the system. The transformation $(n_L, n_R) \leftrightarrow (n, N)$ leads to $P(N, n, T - T_0)$ [26].

Statistics of particle change on the system: Since $\tilde{P}(N) = \sum_n \tilde{P}(N, n)$, the GF, $\tilde{Z}(\Lambda)$, for $\tilde{P}(N)$ satisfies $\frac{1}{(T-T_0)} \left[\frac{\partial \ln \tilde{Z}(\Lambda)}{\partial (i\Lambda)} \right] = 0$, thus $\tilde{Z}(\Lambda, T - T_0) = 1$ and therefore, $\tilde{P}(N, T - T_0) = \delta_{N0}$. This means that fluctuations in the electron change on the system die in the long time limit. This is due to the finite dimensionality of the system Hilbert space.

Statistics of particles exchanged between reservoirs: Generating function $\tilde{Z}(\lambda)$ for the probability distribution of n electrons transferred from the right to the left reservoir, $\tilde{P}(n) = \sum_N \tilde{P}(N, n)$, is obtained as [26]

$$\begin{aligned} \frac{1}{(T-T_0)} \left[\frac{\partial \ln \mathcal{Z}(\lambda, T-T_0)}{\partial (i\lambda)} \right] &= \frac{1}{T_p} \int_0^{T_p} dt \int_{-\infty}^{+\infty} \frac{d\omega}{2\pi} \\ &\left[\frac{\Gamma_L(t) \Gamma_R(t) f(\omega) (1 - f(\omega)) [e^{i\lambda} - e^{-i\lambda}]}{\Delta(\omega, t)} \right. \\ &+ \frac{[\dot{\Gamma}_L(t) + \dot{\Gamma}_R(t)]}{4[\Delta(\omega, t)]^2} (\omega - \omega_0) f'(\omega) \left([\Gamma_R^2(t) - \Gamma_L^2(t)] \right. \\ &+ \Gamma_L(t) \Gamma_R(t) (1 - 2f(\omega)) [e^{i\lambda} - e^{-i\lambda}] \Big) \\ &- \frac{\Gamma_L(t) + \Gamma_R(t)}{2[\Delta(\omega, t)]^2} \left([\Gamma_L(t) \dot{\Gamma}_R(t) - \Gamma_R(t) \dot{\Gamma}_L(t)] f(\omega) (1 - f(\omega)) \right. \\ &\left. \left. \left\{ (0.5 - f(\omega)) + (\omega - \omega_0) f'(\omega) \right\} [e^{i\lambda} + e^{-i\lambda} - 2] \right) \right] \end{aligned} \quad (13)$$

where $f'(\omega) = \frac{\partial f(\omega)}{\partial \omega}$, $\Gamma_\alpha(t) = 2\pi|g_\alpha(t)|^2\rho$, $\dot{\Gamma}_\alpha(t) = \frac{\partial \Gamma_\alpha(t)}{\partial t}$, $\Delta(\omega, t) = (\omega - \omega_0)^2 + (\frac{\Gamma_L(t) + \Gamma_R(t)}{2})^2 + \Gamma_L(t)\Gamma_R(t)f(\omega)(1 - f(\omega))[e^{i\lambda} + e^{-i\lambda} - 2]$, and T_p is the time period of driving which is assumed to be much larger than the internal relaxation time of the system. Here it is assumed that the second measurement is carried out after $q = \frac{T - T_0}{T_p}$ number of cycles of driving (even if it is not the case the error is insignificant at long measurement time, for which the present formalism is developed). The first integral in Eq. (13) is the so-called dynamical contribution (similar to steady-state contribution with parameters replaced with time dependent quantities with a time averaging). The second integral can be converted to parameter integral in (Γ_L, Γ_R) space and is independent of how fast the parameters are varied provided we are in the cyclic adiabatic limit, thus it represents a Berry contribution. When the two drivings are identical $\Gamma_L(t) = \Gamma_R(t)$, the Berry contribution vanishes identically, the area traced in the parameter space (Γ_L, Γ_R) is zero. Equation (13) allows us to compute the full statistics of net particles transferred between the left and right reservoirs. For example, the expression for the average pumped charge is obtained by setting $\lambda = 0$ in Eq. (13). The average number of electrons pumped per cycle is obtained as

$$N_{pump} = \frac{\beta}{8\pi^2} \int_0^{T_p} dt \partial_t (\Gamma_L(t) - \Gamma_R(t)) \Im \Psi^{(1)}(Z) \quad (14)$$

where $Z = \frac{1}{2} + i\frac{\beta}{2\pi}[\mu - \omega_0 - i(\frac{\Gamma_L(t) + \Gamma_R(t)}{2})]$ and $\Im \Psi^{(1)}(Z)$ is the imaginary part of the trigamma function $\Psi^{(1)}(Z)$ of Z [27]. It is clear that $N_{pump} = 0$ when $\omega_0 = \mu$ since $\Im \Psi^{(1)}(Z) = 0$ in this case. This result may be useful in identifying resonance energy, ω_0 , of an unknown quantum system at the junction by applying an external gate voltage on the system such that the net pumped charge is zero. Indeed, also when $\Gamma_L(t) = \Gamma_R(t)$, there is no net pumping of electrons between the two reservoirs. Thus the flux is purely driven due to Berry phase effects. Additionally, we find that the flux changes sign from $\mu > \omega_0$ to $\mu < \omega_0$. Similar behavior in the pumped charge was observed in Ref. [28]. This is demonstrated in the Fig. (1) for sinusoidal drivings. Thus, both the phase difference between the drivings and the detuning act as driving forces that gives rise to a net charge flux. Note that these forces are non-thermodynamic. The Berry phase changes sign as the driving is reversed (the area in parameter space is traced in reverse manner) and, as a consequence, the flux reverses the direction. Note that the dynamical part in Eq. (13) only contributes to the even cumulants. The Berry-phase part, on the other hand, in general, contributes to all the cumulants. However for even-cyclic drivings, *i.e.*, $\Gamma_{L(R)}(-X) = \Gamma_{L(R)}(X)$ and $\Gamma_{L(R)}(t) \equiv \Gamma_{L(R)}(t/T_p \pm \phi/2)$, where ϕ is the phase difference, the Berry part contributes only to the odd cumulants. This fact may be helpful in designing experi-

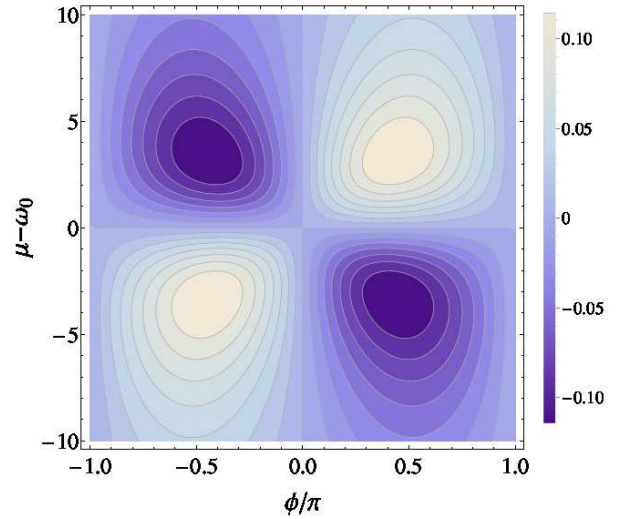


FIG. 1. Average number of particles pumped per cycle, N_{pump} as a function of $\mu - \omega_0$ and ϕ . Here $\Gamma_L(t) = 1 + 10 \sin^2(\frac{\pi t}{T_p} - \frac{\phi}{4})$, $\Gamma_R(t) = 1 + 10 \sin^2(\frac{\pi t}{T_p} + \frac{\phi}{4})$. Parameters Γ_α, μ and ω_0 are in units of β^{-1} .

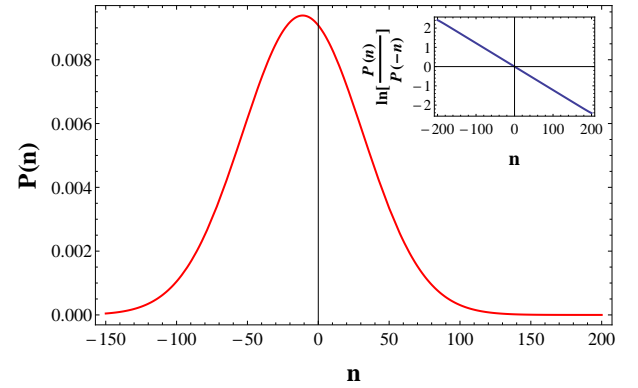


FIG. 2. Probability distribution for the net charge (n) transferred from the right to the left reservoir during the measurement time corresponding to $q = 100$ with $T_p = 100$, $\phi = \pi/2$, $\mu = 0$, and $\omega_0 = 3$. Time is in units of $\hbar\beta$. Drivings are the same as in Fig. (1).

ments to distinguish Berry contribution from the dynamical part. In Fig. (2) we present numerical result for the full distribution function $P(n)$ for measurement time corresponding to $q = 100$. The distribution seems symmetric (second and third cumulants are ~ 18 and ~ 0.05 , respectively) and is peaked around the value $n \sim qN_{pump}$. In the inset we show $\ln(P(n)/P(-n))$ which grows linearly with n , confirming the GC-type fluctuation symmetry. However this symmetry originates due to non-thermodynamic forces and, as a consequence, the GFs for the forward (\mathcal{Z}_F) and backward (\mathcal{Z}_B) drivings satisfy, $\mathcal{Z}_F(\lambda) = \mathcal{Z}_B(-\lambda)$ [26].

We developed an approximation scheme for computing the full counting statistics of adiabatic charge pumps

within the NEGF formalism. We applied this formalism to study the statistics of pumped charge in a single level model where the coupling to the reservoirs are driven adiabatically in time. It is found that a net (non-zero) number of electrons can be transferred between two reservoirs kept at the same thermodynamic states by adiabatically modulating the coupling strengths. An analytic expression is derived for the net charge pumped per cycle entirely due to the geometric (Berry) phase. It is found that

the phase difference between the drivings as well as the energy difference between the level and reservoir chemical potential (at zero bias) are the important parameters that determine the direction of the pumped current. The statistics of the pumped charge is also influenced by the Berry-phase and the corresponding distribution function follows the Gollavati-Cohen type symmetry.

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APPENDIX

DERIVATION OF MEIR-WINGREEN TYPE EXPRESSION FOR GENERATING FUNCTION [EQ.(6) OF THE MAIN TEXT]

Equation (5) of the main text can be recast in terms of the system Greens function matrix alone by substituting the mixed Greens functions in terms of the system Greens functions [22]. We get,

$$\left[\frac{\partial \ln \mathcal{Z}(\lambda_L, \lambda_R, T - T_0)}{\partial(i\lambda_R)} \right] = \int_{T_0}^T dt_1 \int_{T_0}^T dt_2 \text{Tr} \left[\Sigma_R^{+-}(t_1, t_2) G^{+-}(t_2, t_1) - G^{+-}(t_1, t_2) \Sigma_R^{+-}(t_2, t_1) \right] \quad (15)$$

Here the trace is over all the system orbitals and $\Sigma_R^{+-}(t, t')$, etc. are real time projections of self-energy matrix $\Sigma_R(\tau, \tau')$ due to interaction with the right reservoir. It has matrix elements,

$$\Sigma_{R;rr'}(\tau, \tau') = \sum_{k,k'} g_{Rk,r}(\tau) e^{-i(\lambda_R(\tau) - \lambda_R(\tau'))} G_{Rk,Rk'}^0(\tau, \tau') g_{Rk',r'}^*(\tau') \quad (16)$$

where $G_{Rk,Rk'}^0(\tau, \tau')$ being contour ordered Greens functions of free right reservoir). $G^{+-}(t, t')$, etc. are real time projections of system contour ordered Greens function matrix with elements defined in Eq.(7) of main text. Now we use Wigner representation of the quantities in the integrand of Eq.(15) [21] i.e., we use the transformation pair $A(t, t') = \int_{-\infty}^{+\infty} \frac{d\omega}{2\pi} \mathbb{A}(\omega, t_c) e^{i\omega t_q}$ and $\mathbb{A}(\omega, t_c) = \int_{-\infty}^{+\infty} dt_q A(t, t') e^{-i\omega t_q}$ (here $t_c = \frac{t+t'}{2}$ and $t_q = t - t'$ are classical and quantum times respectively) to get

$$\left[\frac{\partial \ln \mathcal{Z}(\lambda_L, \lambda_R, T - T_0)}{\partial(i\lambda_R)} \right] = \int_{T_0}^T dt_c \int_{-(T-T_0)}^{(T-T_0)} dt_q \int_{-\infty}^{+\infty} \frac{d\omega_1}{2\pi} \int_{-\infty}^{+\infty} \frac{d\omega_2}{2\pi} e^{i(\omega_1 - \omega_2)t_q} \text{Tr} \left[\Sigma_R^{+-}(\omega_1, t_c) \mathbb{G}^{+-}(\omega_2, t_c) - \mathbb{G}^{+-}(\omega_1, t_c) \Sigma_R^{+-}(\omega_2, t_c) \right] \quad (17)$$

Here $t_c = \frac{t_1+t_2}{2}$ and $t_q = t_1 - t_2$. We neglect the effect of transients by assuming that the measurement time ' $T - T_0$ ' is large compared to internal relaxation times and driving time period (hence we send the t_q integral from $-\infty$ to ∞). This leads to

$$\left[\frac{\partial \ln \mathcal{Z}(\lambda_L, \lambda_R, T - T_0)}{\partial(i\lambda_R)} \right] = \int_{T_0}^T dt \int_{-\infty}^{+\infty} \frac{d\omega}{2\pi} \text{Tr} \left[\Sigma_R^{+-}(\omega, t) \mathbb{G}^{+-}(\omega, t) - \mathbb{G}^{+-}(\omega, t) \Sigma_R^{+-}(\omega, t) \right]. \quad (18)$$

SYMMETRIZATION OF GENERATING FUNCTION

From $\mathcal{Z}(\lambda_L, \lambda_R, T - T_0)$ we can obtain the combined distribution function $P(n_L, n_R, T - T_0)$ for the electron number change on left and right reservoirs over the measurement time period of $(T - T_0)$ using Eq.(2) of the main text. However in order to compute the statistics of net number (N) of electrons changed on both reservoirs (which is same as the number of electron changed on the system) and the net number (n) of electrons exchanged between both the reservoirs, we perform a coordinate transformation $(n_L, n_R) \rightarrow (N, n) = (\frac{n_L + n_R}{2}, \frac{n_L - n_R}{2})$. Performing this transformation in Eq.(2) of main text, we obtain

$$\tilde{P}(N, n, T - T_0) = \frac{1}{2} \int_0^{2\pi} \frac{d\Lambda}{2\pi} \int_{-2\pi}^{2\pi} \frac{d\lambda}{2\pi} \tilde{\mathcal{Z}}(\Lambda, \lambda, T - T_0) e^{i(\Lambda N + \lambda n)}, \quad (19)$$

where the factor 1/2 appears due to the Jacobian of transformation. $\tilde{\mathcal{Z}}(\Lambda, \lambda, T - T_0)$ is obtained from $\mathcal{Z}(\lambda_L, \lambda_R, T - T_0)$ by performing a coordinate transformation $(\lambda_L, \lambda_R) \rightarrow (\Lambda, \lambda) = (\lambda_L + \lambda_R, \lambda_L - \lambda_R)$. From $\tilde{P}(N, n, T - T_0)$ we can get $\tilde{P}(N, T - T_0)$ or $\tilde{P}(n, T - T_0)$ by summing over 'n' or 'N' respectively.

Summing over N , we get

$$\tilde{P}(n, T - T_0) = \frac{1}{2} \int_{-2\pi}^{2\pi} \frac{d\lambda}{2\pi} \tilde{\mathcal{Z}}(\lambda, T - T_0) e^{i\lambda n} = \int_0^{2\pi} \frac{d\lambda}{2\pi} \tilde{\mathcal{Z}}(\lambda, T - T_0) e^{i\lambda n}, \quad (20)$$

where the second equality follows from the 2π periodicity of $\tilde{\mathcal{Z}}(\lambda, T - T_0)$.

λ -DEPENDENT WIGNER TRANSFORMED GREENS FUNCTIONS FOR THE RESONANT LEVEL MODEL

In this section we describe the procedure to get λ -dependent Greens functions (up to lowest linear order correction in driving) for the resonant level model. We calculate Greens functions on the Keldysh contour which goes from $-\infty$ to ∞ and back to ∞ under large measurement time assumption in order to neglect initial correlations.

The non-interacting Greens function $\check{\mathbb{G}}_0$ is defined below Eq. (8) in the main text. The inverse of $\check{\mathbb{G}}_0$ in Wigner transformed system is obtained as

$$\check{\mathbb{G}}_0^{-1}(\omega, t) = \begin{pmatrix} \omega - \omega_0 + i\eta & 0 \\ 0 & \omega - \omega_0 - i\eta \end{pmatrix}$$

where $\eta = 0^+$. The λ -dependent Wigner transformed self-energy (with terms up to linear in first derivative in classical time) due to coupling to the reservoirs is $\check{\Sigma}(\omega, t) = \sum_{\alpha=L,R} \check{\Sigma}_{\alpha}(\omega, t)$ with

$$\check{\Sigma}_{\alpha}(\omega, t) = \begin{pmatrix} -i\Gamma_{\alpha}(t)(0.5 - f_{\alpha}(\omega)) & i\Gamma_{\alpha}(t)f_{\alpha}(\omega)e^{i\lambda_{\alpha}} \\ i\Gamma_{\alpha}(t)(1 - f_{\alpha}(\omega))e^{-i\lambda_{\alpha}} & i\Gamma_{\alpha}(t)(0.5 - f_{\alpha}(\omega)) \end{pmatrix}$$

Here we assume that the system-reservoir couplings are real and independent of energy (wide-band approximation). The first order correction to the self-energy due to external driving is then zero.

Using Eq.(11) of the main text together with the above two equations for $\check{\mathbb{G}}_0^{-1}$ and $\check{\Sigma}_{\alpha}$, we get an expression for the adiabatic Greens function as

$$\check{\mathbb{G}}_{ad}(\omega, t) = \frac{1}{\Delta(\omega, t)} \begin{pmatrix} \omega - \omega_0 - i\Gamma_L(t)(0.5 - f_L(\omega)) - i\Gamma_R(t)(0.5 - f_R(\omega)) & i\Gamma_L(t)f_L(\omega)e^{i\lambda_L} + i\Gamma_R(t)f_R(\omega)e^{i\lambda_R} \\ i\Gamma_L(t)(1 - f_L(\omega))e^{-i\lambda_L} + i\Gamma_R(t)(1 - f_R(\omega))e^{-i\lambda_R} & \omega - \omega_0 + i\Gamma_L(t)(0.5 - f_L(\omega)) + i\Gamma_R(t)(0.5 - f_R(\omega)) \end{pmatrix}$$

with

$$\Delta(\omega, t) = (\omega - \epsilon)^2 + \left(\frac{\Gamma_L(t) + \Gamma_R(t)}{2} \right)^2 + \Gamma_L(t)\Gamma_R(t)[f_L(\omega)(1 - f_R(\omega))(e^{i(\lambda_L - \lambda_R)} - 1) + f_R(\omega)(1 - f_L(\omega))(e^{-i(\lambda_L - \lambda_R)} - 1)]$$

where $f_L(\omega)$ and $f_R(\omega)$ are Fermi functions of the left and right reservoirs, respectively.

Using $\check{\mathbb{G}}_{ad}(\omega, t)$ in Eq.(12) of main text we calculate the lowest order correction to the Greens functions. We give expressions only for '+'-' and '-+' components.

$$\begin{aligned} \check{\mathbb{G}}^{+-}(\omega, t) &= \frac{i\Gamma_L(t)f_L(\omega)e^{i\lambda_L} + i\Gamma_R(t)f_R(\omega)e^{i\lambda_R}}{\Delta(\omega, t)} - \frac{i[\dot{\Gamma}_L + \dot{\Gamma}_R](\omega - \omega_0)}{2[\Delta(\omega, t)]^2} [\Gamma_L f'_L(\omega)e^{i\lambda_L} + \Gamma_R f'_R(\omega)e^{i\lambda_R}] \\ &\quad + \frac{i(\Gamma_L(t)\dot{\Gamma}_R(t) - \Gamma_R(t)\dot{\Gamma}_L(t))}{2[\Delta(\omega, t)]^2} [(1 - 2f_R(\omega))f_L(\omega)e^{i\lambda_L} - (1 - 2f_L(\omega))f_R(\omega)e^{i\lambda_R}] \\ &\quad + \frac{i(\omega - \omega_0)}{[\Delta(\omega, t)]^2} [\Gamma_L(t)\dot{\Gamma}_R(t)f_R(\omega)f'_L(\omega) - \Gamma_R(t)\dot{\Gamma}_L(t)f_L(\omega)f'_R(\omega)](e^{i\lambda_L} - e^{i\lambda_R}) \end{aligned} \quad (21)$$

$$\begin{aligned} \check{\mathbb{G}}^{-+}(\omega, t) &= \frac{i\Gamma_L(t)(1 - f_L(\omega))e^{-i\lambda_L} + i\Gamma_R(t)(1 - f_R(\omega))e^{-i\lambda_R}}{\Delta(\omega, t)} + \frac{i[\dot{\Gamma}_L + \dot{\Gamma}_R](\omega - \omega_0)}{2[\Delta(\omega, t)]^2} [\Gamma_L f'_L(\omega)e^{-i\lambda_L} + \Gamma_R f'_R(\omega)e^{-i\lambda_R}] \\ &\quad - \frac{i(\Gamma_L(t)\dot{\Gamma}_R(t) - \Gamma_R(t)\dot{\Gamma}_L(t))}{2[\Delta(\omega, t)]^2} [(1 - 2f_R(\omega))(1 - f_L(\omega))e^{-i\lambda_L} - (1 - 2f_L(\omega))(1 - f_R(\omega))e^{-i\lambda_R}] \\ &\quad - \frac{i(\omega - \omega_0)}{[\Delta(\omega, t)]^2} [\Gamma_L(t)\dot{\Gamma}_R(t)(1 - f_R(\omega))f'_L(\omega) - \Gamma_R(t)\dot{\Gamma}_L(t)(1 - f_L(\omega))f'_R(\omega)](e^{-i\lambda_L} - e^{-i\lambda_R}) \end{aligned} \quad (22)$$

Using $\check{\mathbb{G}}^{+-}(\omega, t)$, $\check{\mathbb{G}}^{-+}(\omega, t)$, $\check{\Sigma}_R^{+-}(\omega, t)$ and $\check{\Sigma}_R^{-+}(\omega, t)$ in Eq.(18) we get an expression for $\frac{1}{(T-T_0)} \left[\frac{\partial \ln \mathcal{Z}(\lambda_L, \lambda_R, T-T_0)}{\partial(i\lambda_R)} \right]$ to lowest order correction,

$$\begin{aligned} & \frac{1}{(T-T_0)} \left[\frac{\partial \ln \mathcal{Z}(\lambda_L, \lambda_R, T-T_0)}{\partial(i\lambda_R)} \right] = \\ & \frac{1}{T_p} \int_0^{T_p} dt \int_{-\infty}^{+\infty} \frac{d\omega}{2\pi} \frac{\Gamma_L(t)\Gamma_R(t)}{\Delta(\omega, t)} [f_R(\omega)(1-f_L(\omega))e^{-i(\lambda_L-\lambda_R)} - f_L(\omega)(1-f_R(\omega))e^{i(\lambda_L-\lambda_R)}] \\ & + \frac{1}{T_p} \int_0^{T_p} dt \int_{-\infty}^{+\infty} \frac{d\omega}{2\pi} \frac{1}{[\Delta(\omega, t)]^2} \times \\ & \left[\frac{\Gamma_R(t)[\Gamma_L(t)\dot{\Gamma}_R(t) - \Gamma_R(t)\dot{\Gamma}_L(t)]}{2} [f_L(\omega) - f_R(\omega)] - \frac{\Gamma_R(t)[\dot{\Gamma}_L(t) + \dot{\Gamma}_R(t)](\omega - \omega_0)}{2} [\Gamma_L(t)f'_L(\omega) + \Gamma_R(t)f'_R(\omega)] \right. \\ & + \frac{\Gamma_R(t)[\Gamma_L(t)\dot{\Gamma}_R(t) - \Gamma_R(t)\dot{\Gamma}_L(t)]}{2} (1 - 2f_R(\omega)) [f_L(\omega)(1-f_R(\omega))(e^{i(\lambda_L-\lambda_R)} - 1) + f_R(\omega)(1-f_L(\omega))(e^{-i(\lambda_L-\lambda_R)} - 1)] \\ & - \frac{\Gamma_L(t)\Gamma_R(t)[\dot{\Gamma}_L(t) + \dot{\Gamma}_R(t)](\omega - \omega_0)}{2} f'_L(\omega) [(1-f_R(\omega))(e^{i(\lambda_L-\lambda_R)} - 1) + f_R(\omega)(e^{-i(\lambda_L-\lambda_R)} - 1)] \\ & + \Gamma_L(t)\Gamma_R(t)\dot{\Gamma}_R(t)(\omega - \epsilon) f'_L(\omega) f_R(\omega)(1-f_R(\omega)) [e^{i(\lambda_L-\lambda_R)} + e^{-i(\lambda_L-\lambda_R)} - 2] \\ & \left. - \Gamma_R^2(t)\dot{\Gamma}_L(t)f'_R(\omega)(\omega - \omega_0) [f_L(\omega)(1-f_R(\omega))(e^{i(\lambda_L-\lambda_R)} - 1) + f_R(\omega)(1-f_L(\omega))(e^{-i(\lambda_L-\lambda_R)} - 1)] \right] \end{aligned} \quad (23)$$

where $f'_\alpha(\omega)$ is ω derivative of $f_\alpha(\omega)$, $\dot{\Gamma}_\alpha(t)$ is time derivative of $\Gamma_\alpha(t)$ and ' T_p ' is the time period of driving which is assumed to be much larger than the internal relaxation time of the system. A similar expression for $\frac{1}{(T-T_0)} \left[\frac{\partial \ln \mathcal{Z}(\lambda_L, \lambda_R, T-T_0)}{\partial(i\lambda_L)} \right]$ can be obtained by swapping 'L' and 'R' labels in Eq. (23). Here it is assumed that the second measurement is carried out after an integer number of cycles $n = \frac{T-T_0}{T_p}$ of driving has been performed (even if it is not the case the error is minimal for large time statistics, for which the present formalism is developed). Using expressions for $\frac{1}{(T-T_0)} \left[\frac{\partial \ln \mathcal{Z}(\lambda_L, \lambda_R, T-T_0)}{\partial(i\lambda_R)} \right]$ and $\frac{1}{(T-T_0)} \left[\frac{\partial \ln \mathcal{Z}(\lambda_L, \lambda_R, T-T_0)}{\partial(i\lambda_L)} \right]$, expression for the generating function for particle change on system and particle exchanged between reservoirs can be obtained. These expressions are presented in the main text for $f_L(\omega) = f_R(\omega) = f(\omega)$ case.

SYMMETRIES OF THE GREENS FUNCTIONS

As the full system is evolving with respect to a Hamiltonian, the evolution is unitary when $\lambda_L = \lambda_R = 0$. As a consequence, matrix elements of $\check{\mathbb{G}}(\tau, \tau')$ satisfy the symmetries [21]: (i) $[\check{\mathbb{G}}_{mn}^{++}(t, t')]^* = \check{\mathbb{G}}_{nm}^{--}(t', t)$, (ii) $[\check{\mathbb{G}}_{mn}^{--}(t, t')]^* = \check{\mathbb{G}}_{nm}^{++}(t', t)$, (iii) $[\check{\mathbb{G}}_{mn}^{+-}(t, t')]^* = -\check{\mathbb{G}}_{nm}^{+-}(t', t)$ and (iv) $[\check{\mathbb{G}}_{mn}^{-+}(t, t')]^* = -\check{\mathbb{G}}_{nm}^{-+}(t', t)$. These symmetries in Wigner representation can be summarized in a matrix form as

$$\check{\mathbb{G}}(\omega, \mathbf{t}) = \begin{bmatrix} \check{\mathbb{G}}^{++}(\omega, t) & \check{\mathbb{G}}^{+-}(\omega, t) \\ \check{\mathbb{G}}^{-+}(\omega, t) & \check{\mathbb{G}}^{--}(\omega, t) \end{bmatrix} \implies [\check{\mathbb{G}}(\omega, \mathbf{t})]^* = \begin{bmatrix} [\check{\mathbb{G}}^{--}(\omega, t)]^T & -[\check{\mathbb{G}}^{+-}(\omega, t)]^T \\ -[\check{\mathbb{G}}^{-+}(\omega, t)]^T & [\check{\mathbb{G}}^{++}(\omega, t)]^T \end{bmatrix}$$

The approximate Greens function matrix used in this work can be expressed as

$$\check{\mathbb{G}}(\omega, t) = \check{\mathbb{G}}_{ad}(\omega, t) + \frac{i}{2} \left[\{ \partial_\omega \check{\mathbb{G}}_{ad}(\omega, t) \} \check{\mathbb{G}}_{ad}^{-1}(\omega, t) \partial_t \check{\mathbb{G}}_{ad}(\omega, t) - \{ \partial_t \check{\mathbb{G}}_{ad}(\omega, t) \} \check{\mathbb{G}}_{ad}^{-1}(\omega, t) \partial_\omega \check{\mathbb{G}}_{ad}(\omega, t) \right] \quad (24)$$

This approximate Greens function satisfies all the above symmetries provided $\check{\mathbb{G}}_{ad}(\omega, t)$ satisfies the above symmetries which in turn requires $\check{\mathbb{G}}_0(\omega, t)$ and $\check{\Sigma}(\omega, \mathbf{t})$ to satisfy the above symmetries, which they do. Another important symmetry is $\check{\mathbb{G}}_{mn}^{++}(t, t') - \check{\mathbb{G}}_{mn}^{--}(t, t') = \check{\mathbb{G}}_{mn}^{+-}(t, t') - \check{\mathbb{G}}_{mn}^{-+}(t, t')$ which in Wigner representation becomes $\check{\mathbb{G}}_{mn}^{++}(\omega, t) - \check{\mathbb{G}}_{mn}^{--}(\omega, t) = \check{\mathbb{G}}_{mn}^{+-}(\omega, t) - \check{\mathbb{G}}_{mn}^{-+}(\omega, t)$ is also satisfied by the above approximate Greens function provided $\check{\mathbb{G}}_0(\omega, t)$ and $\check{\Sigma}(\omega, t)$ also satisfy it, which they do.

DETAILED FLUCTUATION THEOREM FOR PUMPED CHARGE

We consider a special case when both the reservoirs are at the same thermodynamic states (i.e., $\beta_L = \beta_R = \beta$ and $\mu_L = \mu_R = \mu$) and drivings are of the form $\Gamma_L(t) = \Gamma(\frac{2\pi t}{T_p} - \frac{\phi}{2})$ and $\Gamma_R(t) = \Gamma(\frac{2\pi t}{T_p} + \frac{\phi}{2})$ (where $\Gamma(x)$ is an even periodic function with period 2π). During the time-reversed evolution, the drivings at time t are obtained by substituting $t \rightarrow T_p - t$ for drivings in the forward evolution. Clearly the Hamiltonian does not have time-reversal symmetry as $\Gamma_{L(R)}(T_p - t) = \Gamma_{R(L)}(t)$, the left and right couplings switch roles. Then using Eq. (13) of the main text, it can be shown that, $\tilde{Z}_{\mathcal{F}}(\lambda, T - T_0) = \tilde{Z}_{\mathcal{B}}(-\lambda, T - T_0)$, where $\tilde{Z}_{\mathcal{F}}$ and $\tilde{Z}_{\mathcal{B}}$ are moment generating functions of probability distribution function for number of particles exchanged between two reservoirs with forward and backward driving protocols, respectively. This is the Gallavotti-Cohen type symmetry for zero bias case. This symmetry leads to a detailed fluctuation theorem (FT)

$$\lim_{(T-T_0) \rightarrow \infty} \frac{P_F(n, T - T_0)}{P_B(-n, T - T_0)} = 1, \quad (25)$$

which is consistent with the standard (non-driven) steady-state FT for charge transfer in single resonant level system at equilibrium (zero external bias). Result in Eq. (25) is a consequence of the $\mathcal{P} \otimes \Theta$, where \mathcal{P} and Θ are the parity (left \leftrightarrow right) and the time-reversal operations, symmetry of the Hamiltonian at zero bias and the drivings considered above. However, at steady-state $P_F \equiv P_B$, and the above relation leads to $P_F(n) = P_F(-n)$ at large measurement times. However, for a driven case, as we show in the main text, we find that $\tilde{Z}_{\mathcal{F}}(\lambda, T - T_0) \neq \tilde{Z}_{\mathcal{F}}(-\lambda, T - T_0)$ due to Berry phase which leads to a finite net charge transfer between reservoirs .

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